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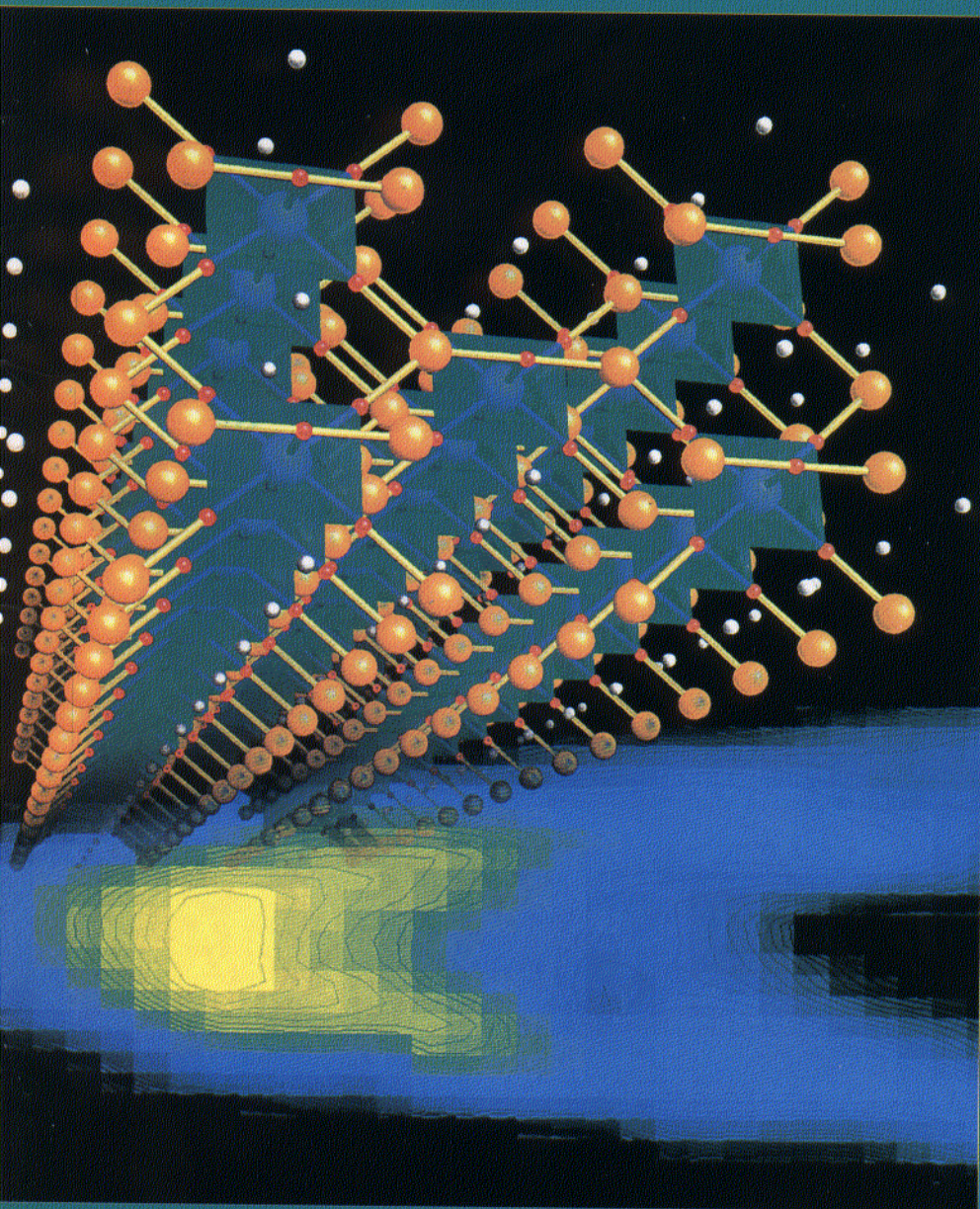
Haldane Spin Chains in a Staggered Field: 2-1-1-5 Rare Earth Nickelates

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Recent neutron scattering studies show that R_2BaNiO_5 (R = rare earth) quasi-1-D antiferromagnets are ideal model systems for studying the behavior of Haldane spin chains in strong *staggered* magnetic fields.

The novel quantum-disordered ground state and the famous Haldane gap in the magnetic excitation spectrum [1] have kept $S=1$ one-dimensional (1-D) Heisenberg antiferromagnets (AF) at the center of attention of condensed matter physicists for almost 15 years. Among the more recent developments are studies of such systems in external magnetic fields (a comprehensive bibliography can be found in Refs. [2]). The effect of a *staggered* field H_π, to which the Haldane chain is

Figure 1. *Measured static staggered moment on the Ni-chains in several R_2BaNiO_5 compounds (symbols) plotted as a function of the rare earth moment (bottom axes) or the effective staggered field (top axis). The solid line is a 1-parameter theoretical fit. The data shown are from Refs. [8,9,11].*

most susceptible, is expected to be no less dramatic. In the past this problem was not given much attention in literature, as the required conditions were thought to be almost impossible to realize in an experiment [3]. One clever trick to obtain a field oscillating on the *microscopic* scale is to make use of an intrinsic magnetic modulation in a material that, in addition to integer-spin Heisenberg chains, has other magnetic ions. Should the latter become ordered magnetically with an appropriate propagation vector, they will project an effective staggered exchange field on the Haldane spin chains. A neat realization of this mechanism was recently found in a class of linear-chain rare earth nickelates with a general formula R_2BaNiO_5. Central to understanding these compounds was the idea that, thanks to the strong in-chain coupling, the Haldane singlet ground state of individual $S=1$ Ni-chains is *not* destroyed by long-range magnetic ordering. For the first time the influence of a strong staggered field on the properties of $S=1$ quantum spin chains, and particularly on the triplet Haldane-gap excitations, could be studied experimentally.

Neutron scattering experiments have been crucial to these studies. The first inelastic measurements on Pr_2BaNiO_5 and $(Nd_xY_{1-x})_2BaNiO_5$ were done at the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory. Because of the current shutdown of the HFBR, the project has gradually evolved into a multi-national collaboration involving BNL, Aoyama-Gakuin University (Tokyo), ORNL, NIST, CEA Grenoble, ILL, University of Delaware, Johns-Hopkins University, Tohoku University, and KEK. With time, a consistent understanding of the physics involved has emerged. The present paper is a review of the recent experimental and theoretical findings.

The most prominent structural feature of R_2BaNiO_5 compounds are $S=1$ Ni^{2+} chains running along the a axis of the orthorhombic crystal structure (see cover illustration). In-chain interactions between the Ni, spins are antiferromagnetic, with the exchange constant $J{\approx}300$ K. Direct interchain coupling is negligible. In fact, the isostructural Y_2BaNiO_5, where the magnetic rare earths are replaced by non-magnetic Y^{3+}, is an excellent example of a 1-D Heisenberg AF: it shows no long-range magnetic order even at low temperatures, and has a Haldane gap $\Delta{\approx}10$ meV in the excitation spectrum [4,5].
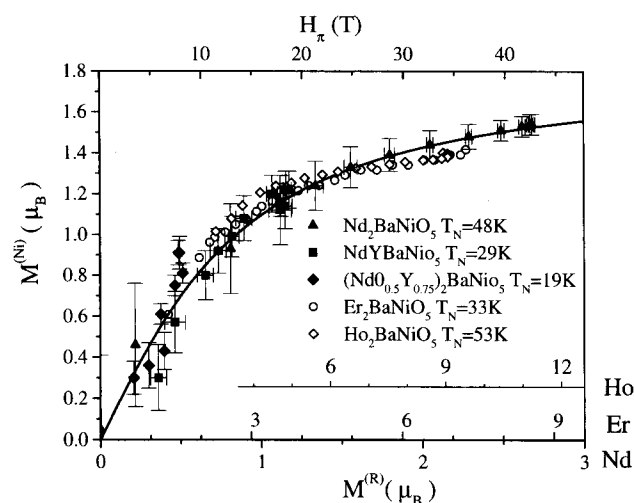
Despite the lack of static order, dynamic AF spin correlations in the Ni-chains remain important at low temperature, decaying over a distance of about six lattice spacings. In R_2BaNiO_5 the magnetic R^{3+} ions are positioned in between the Ni chains. It is through these magnetic sites that interchain interactions are established, giving rise to long-range AF Néel order with transition temperatures ranging from 16 to 80 K [6,7] and static magnetization on both Ni^{2+} and R^{3+}.

First, let us consider static magnetic properties and, in particular, the temperature dependence of sublattice magnetizations. The crucial thing to always keep in mind is that, thanks to strong in-chain interactions at the ordering temperature, significant in-chain spin correlations are already well established. One can therefore view the static staggered moment on the originally disordered Ni chains as being *induced* by the ordered rare earth subsystem. The response of the chains to this effective staggered exchange field is described by a *staggered magnetization curve* $M_\pi(H_\pi)$ of an isolated $S=1$ quantum AF. This function is one of the principal characteristics of the Haldane state, and can be expected to be a universal property of all R_2BaNiO_5 compounds: it is entirely determined by in-chain Ni-Ni coupling and is therefore independent of the type of rare earth involved. The first experimental measurement $M_\pi(H_\pi)$ was obtained [8,9] by analyzing the results of rather straightforward neutron diffraction experiments on R_2BaNiO_5 powders ($R=(Nd_xY_{1-x})$ [6,7,10], $R=Er$ and $R=Ho$ [11]). Assuming that H_π is proportional to the magnetization on the rare earth sites, M is recovered by simply plotting the measured magnetization of the Ni sublattice versus that of the R-subsystem, as shown in Figure 1. The scaling of the abscissa for different rare earth species is arbitrary, yet the apparent data collapse for all $(Nd_xY_{1-x})_2BaNiO_5$ compounds, where the Néel temperatures range from 20 to 50 K, is quite encouraging.

Of course, the coupling between the Ni and R sublattices needs to be treated self consistently. This was accomplished by constructing a simple mean field (MF) model that incorporates the independently measured function M for the Ni spins and classical Brillouin functions for the rare earth moments. From the MF analysis, the actual magnitudes of staggered fields involved were extracted [8,9], and abscissa in Figure 1 could thus be re-scaled to show the staggered field in appropriate units (Figure 1, top axis). Staggered fields of up to *40* T are found! Recent analytical [12,9] (solid line in Figure 1) and numerical [13] predictions for isolated quantum chains are in excellent agreement with these experimental results.
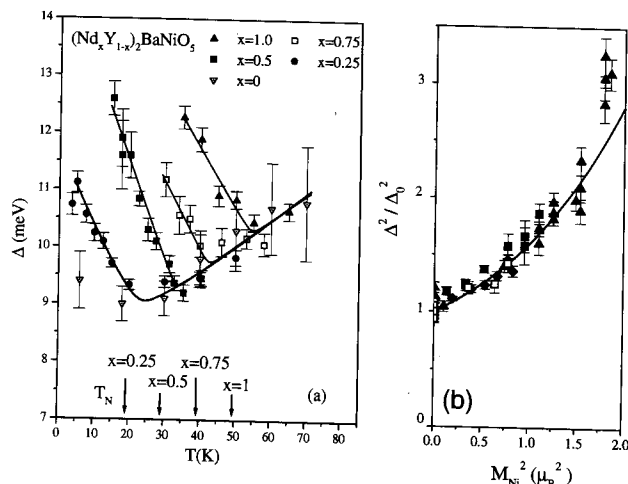

Figure 2. *(a) Measured temperature dependence of the Haldane gap energy in several $(Nd_xY_{1-x})_2BaNiO_5$ compounds with different ordering temperatures. Solid lines are guides for the eye. (b) Same data plotted as a function of the staggered moment induced in the Ni-chains. The solid line is a parameter-free theoretical prediction [12]. The experimental data are taken from Refs. [10,9].*

As far as the spin dynamics are concerned, the most striking observation is that Haldane gap spin excitations are found *both above and below the Néel temperature* in all R_2BaNiO_5 materials studied so far [14,15,10,9]. These excitations are virtually indistinguishable from the Haldane modes in Y_2BaNiO_5. At $T>T_N$ they occur at exactly the same energy and are well characterized by the dynamic structure factor calculated for an isolated quantum $S=1$ chain. The only thing that changes upon cooling through the Néel point is the gap energy Δ. In the ordered state, Δ *increases* with decreasing T [Figure 2(a)]. In all R_2BaNiO_5 species studied to date, this change in Δ is *universally* related to the magnitude of the induced moment on the Ni-sublattice, as illustrated in Figure 2(b) for Nd-Y systems.

It is well known that a number of integer-spin quasi-1D materials that undergo long-range ordering at low temperature feature distinct Haldane-gap modes in the paramagnetic phase, $CsNiCl_3$ being a classic example [16]. In the latter system however, as the Néel temperature is approached from above, the Haldane modes develop a pronounced transverse dispersion and undergo a complete softening at the 3-D AF zone-center, transforming into conventional gapless spin waves in the ordered state. Not so in R_2BaNiO_5 compounds! Here the Haldane excitations do not show any sign of softening or transverse dispersion at any wave vector or temperature [14,17]. Conventional spin waves (order-
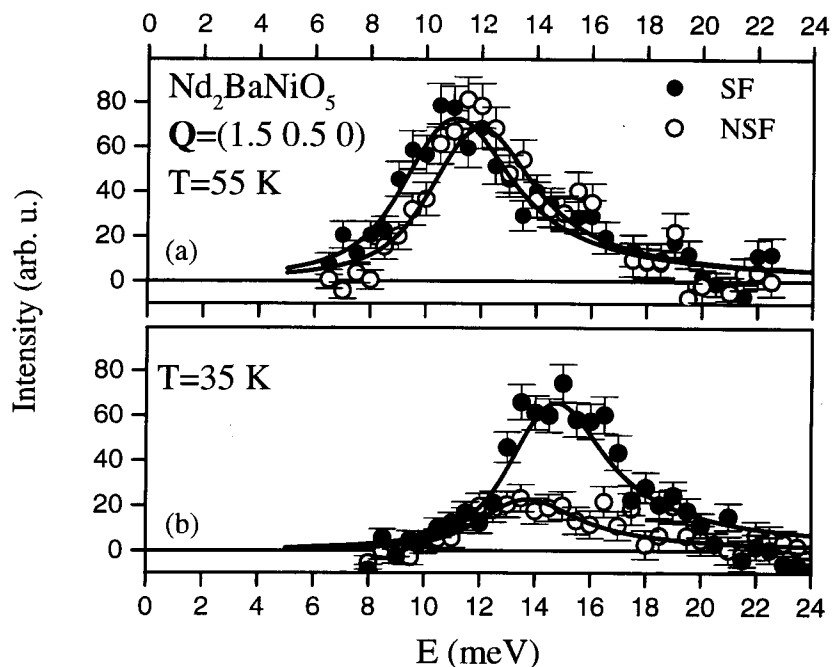
Figure 3. *Polarized-neutron inelastic scans measured in Nd$_2$BaNiO$_5$ at the 1-D AF zone center above (a) and below (b) the Néel temperature. Open and solid circles correspond to the longitudinal and transverse Haldane-gap excitations, respectively. The data are from Ref. [19].*

parameter excitations) appear in the ordered phase as a separate entity and coexist with Haldane-gap modes [14,9].

At first the observed increase of the gap energy came as a surprise. Very soon, however, such behavior was theoretically predicted for isolated Haldane chains in a static staggered field [18,12,13]. For R_2BaNiO$_5$ the situation is of course more complex: the interactions between R and Ni-subsystems may lead to a mixing between Haldane modes and conventional spin waves. This mixing in general can be modeled using the Random Phase Approximation, essentially a dynamic version of the MF approach described above. However, all the relevant exchange constants and even the actual electronic configurations of the rare earths ions involved are not known with sufficient accuracy for this analysis. It is rather fortunate then that for one particular material, namely Nd$_2$BaNiO$_5$, this information is apparently not required. In this system the conventional spin waves that appear below T_N are totally dispersionless and can be seen as cluster modes centered on the Nd^{3+} sites [9]. In this case, the two *transverse*-polarized members of the Haldane triplet do not mix with the order-parameter excitations. They can therefore be regarded as Haldane excitations propagating in *isolated*

$S=1$-chains immersed in an effective *static* staggered field. The temperature dependence of the Haldane gap energy Δ measured in Nd$_2$BaNiO$_5$ could thus be directly compared to theoretical predictions for isolated chains. The solid line in Figure 2(b) is the theoretical result for transverse gap modes [12] plotted over the experimental data points without any adjustable parameters.

Most interesting is the behavior of the longitudinal Haldane excitation, i.e., the one polarized *along* the direction of ordered moments on the Ni-sites. Such an excitation would be absent in any classical spin wave theory, and is a signature of singlet quantum chains. For a long time the longitudinal mode remained elusive. Only recently it was observed for the first time in Nd$_2$BaNiO$_5$ using spin-polarized inelastic neutron scattering [19]. The polarization setup was configured to detect only the longitudinal mode in the non-spin-flip configuration, and only one of the transverse modes in the spin-flip channel. At $T>T_N$, both excitations have similar intensities and appear at almost the same energy [Figure 3(a)]. Below the ordering temperature, the transverse spin gap increases slightly more rapidly than the longitudinal one [Figure 3(b)]. The longitudinal mode becomes weakened, but is still clearly observed in a wide temperature range below T_N.

In summary, one can hope that the basic physical principles that govern the magnetic properties of R_2BaNiO_5 materials are finally understood. The suggested RPA model for Ni-R coupling seems to be a quite adequate theoretical model for these remarkable quantum magnets. Of course, this study is far from being completed. Just one example of a very promising direction for future work is a detailed investigation of the mixing between acoustic spin waves and Haldane-gap excitations. These effects should be particularly important in systems like Pr_2BaNiO_5, where the order-parameter excitations are themselves highly dispersive [14].

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